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REVIEW OF SHORT WAVELENGTH LASERS

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Review of Short Wavelength Lasers

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There has recently been a substantial amount of research devoted to the development of short wavelength amplifiers and lasers. A number of experimental results have been published wherein the observation of significant gain has been claimed on transitions in the EUV and soft X-ray regimes. The present review is intended to discuss the main approaches to the creation of population inversions and laser media in the short wavelength regime, and hopefully aid workers in the field by helping to provide access to a growing literature.

The approaches to pumping EUV and soft X-ray lasers are discussed according to inversion mechanism. The approaches may be divided into roughly seven catagories, including collisional excitation pumping, recombination pumping, direct photoionization and photoexcitation pumping, metastable state storage plus optical pumping, charge exchange pumping, and finally, the extension of free electron laser techniques into the EUV and soft X-ray regimes.

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I. Introduction

During the last two or three years substantial progress has been made in the area of EUV and soft X-ray lasers. From the early report of Ilyukhin et al (1977) in which lasing in neon-like calcium at 600 Å was claimed (Ref. 1), we have seen reported five gain lengths at 182 Å in a laser-heated thin carbon filament (Jacoby et al (1981), Ref. 2), 3.5 gain lengths (and an unpublished claim of even higher) at 182 Å in a magnetically confined solenoidal plasma (Suckewer et al (1984), Ref. 3), and 6.5 gain lengths at 206 and 209 Å in a laser heated thin-film selenium on formvar exploding foil (Matthews et al (1984), Ref. 4). In coming years we might expect many more reports of higher gains attained at increasingly shorter wavelengths, as years' worth of work at many laboratories around the world begins to pay off. This is a very exciting time in the field, and in the present review we shall summarize briefly some of the research which has recently been completed, is ongoing, and may come in future years.

The field of EUV and soft X-ray lasers has been reviewed frequently (Ref. 5-16) in the past, and our primary purpose here is to provide a survey reference list of literature in the field with minor commentary. Our hope is to aid researchers in the various short wavelength laser areas, as well as to provide material from which an interested reader might be able to have somewhat easier access to a sizable literature within the field. The total number of papers in the field is probably in excess of 500 by now, and as we shall cite only a fraction of that number, we can make no claims regarding completeness.

There are several mechanisms which have been proposed to pump EUV and soft X-ray lasers, including

1) Electron collisional excitation.
2) Recombination, with or without rapid cooling.
3) Photoionization of inner-shell electrons.
4) Photoexcitation using resonant line radiation.
5) Storage of excitation in metasable states, and optical laser assist.
6) Charge exchange.
7) Free-electron X-ray laser action.
8) Other approaches.

We shall consider the various approaches in this order in our review. Our purpose is not to make any statements as to which approach is better or worse than which other approach, since it is probable that all will ultimately be successful, rather to provide access to relevant literature.

II. Electron Collisional Excitation

The earliest report of an electron collisional excitation scheme in the EUV which we have available is an unpublished memo of Duguay dating back to 1969 (Ref. 17). The proposal involves inversion of $3p-3s$ transitions in F-like cobalt pumped by electron impact excitation from ground state fluorine-like ions. The scheme was proposed as the extension of the analog optical $3p-3s$ Ne II lasers [18] into the EUV and soft X-ray regimes. The basic idea is that 2p electrons can be excited either directly or otherwise to the 3p shell, and due to the fast radiative decay of singly excited states with a 3s electron, a population inversion can be maintained in a quasi-CW sense on $3p-3s$ transitions. The earliest published report of this approach which we have found is in the review paper of Molchanov (1972) [13], in which a discussion is given concerning the extension of optical 3p-3s lasers in the lithium, beryllium, boron, and nitrogen sequences into the UV regions. In 1973, Duguay discussed 3p-3s transitions in the nitrogen sequence, and the extension of the approach into the X-ray regime [7].

Other electron collisional excitation schemes were discussed early on in the literature [19–22]; however, the $3p-3s$ approach has more or less dominated the area, and seems nowadays to be synonymous with electron collisional excitation schemes.

The first quantitative analysis of the kinetics and gain on $3p-3s$ transitions which we know of is an NRL report of 1974 [23], which was published later in Ref. 24. Subsequent calculations include those of Whitney and Davis (1975) [25] (carbon-like), Zherikhin et al. (1976) [26] (neon-like), Vinogradov et al (1977) [27] (neon-like), Palumbo and Elton (1977) [28] (carbon-like and helium-like), and Davis et al (1977) [29] (carbon-like).

The first positive experimental results were published by Ilyukhin et al (1978) [1], who claimed to have observed 10 nanojoules of laser light near 600 Å on their diagnostic film, and inferred a total output of a microjoule from calcium $3p-3s$ transitions. The investigators pumped a calcium target with a neodymium laser with pulse lengths in the 2.5-5 nsec range and pulse energy of 30 Joules. A laser cavity was constructed with total length 14 cm, and the calcium plasma had a length of 0.15 mm. The laser intensity on the calcium target was varied in the range of 2 - 7.5 x 10^{10} Watts/cm. Elton (1978) [9] reported that collimation to approximately 2 mrad was obtained [30]

There are no reports of this experiment being reproduced elsewhere as far as we know. In retrospect it is extremely unfortunate that no precise wavelengths were determined and reported from the experiment, as in post-analysis it may have been possible to tell whether the laser lines were due to monopole excitation as according to theory, or due to other mechanisms. There is controversy even years after as to whether a laser was actually demonstrated, especially considering comments in Ref. 15, which we quote here: "The results of this experiment are promising, but it is still too early to talk about a laser device."

Subsequent theoretical work has focused almost entirely on the neon-like sequence, although it is true that in an experiment it is much easier to produce a plasma consisting of about half neon-like ions as opposed to half carbon-like (or other L-shell sequence) ions In Ref. 31-33, the Lebedev group continued their analysis of neon-like ions, including effects due to intermediate coupling between the M-shell levels and estimating trapping contributions from the L-M resonance trasitions. Theoretically, the maximum $3p-3s$ gain obtainable is low for low Z elements (0.4-0.5 cm^{-1} for S VII) and scales like $Z^{4.5}$ to mid Z elements (20-40 cm^{-1} for Fe XVII) [33]. Substantial gain was calculated also for $3d-3p$ transitions between states with $2s$ holes.

At this point some comment is in order regarding excitation mechanism. The original proposals of $3p-3s$ inversions in the EUV and soft X-ray regimes were not overly concerned with the precise excitation mechanisms; however, the early analyses focused on the importance of electron monopole excitation in the creation of a population inversion [23,24,27]. In the neon-like sequence, monopole excitation would tend to populate $3p$ levels with total angular momentum $J = 0$, and as only $3s$ levels with $J = 1$ decay rapidly spontaneously, one would expect monopole excitation to lead to high gain J=0 to J=1 $3p-3s$ transitions. In Ref. 32 and 33 substantial gain was computed for transitions from starting from $J = 2$ $3p$ states (in addition to the $J = 0$ states), which are not populated by direct monopole excitation, and which are not pumped primarily by quadrupole excitation from the ground state as the relevant excitation cross sections are not so large.

In Ref. 34, the focus was on gain on the principal $J = 0$ transition, and in Ref. 35, results were presented for both $J = 0$ and $J = 2$ lines although in krypton the classical monopole excitation path leads to the largest theoretical gain. Ref. 36 is concerned with monopole excitation as well. The design and analysis of Ref. 12 was focused on the principal 0-1 transition, although gains on all other lines were monitored and were found theoretically to be less.

Recently, in a well-diagnosed set of experiments at LLNL, significant amplification (6.5 gain-lengths) was reported [4] on the 206 and 209 Å lines of neon-like selenium from a laser-pumped thin film exploding foil target [37], which are lines originating from $J = 2$ $3p$ states. Substantial gain was also found on the analog transitions of yttrium at 155 and 157 Å. No significant amplification was observed on lines with $J = 0$ $3p$ states. In selenium, theoretically the 0-1 line near 182 Å was predicted to have the highest gain, and the 169

Å line, also a 0-1 line, should have shown some amplification [37]. The lines originating from J = 2 $3p$ transitions are in very reasonable agreement with the experimental results showing highest gain, and many other parameters (electron temperature, density) are in good agreement with theory. There was speculation that the dominant 0-1 line may have been resonantly absorbed through an unfortunate resonance; however, this seems dubious as similar non-amplification was found in Ge, Br and Y.

The fact that up to 6.5 gain-lengths were obtained experimentally is a remarkable result, and in the coming year further experiments will push the gain sufficiently high (12-14 gain-lengths) to saturate the transitions. The fact that no amplification was found on the 0-1 transitions is not currently understood theoretically. There were other discrepences between theory and experiment which will receive further discussion in the literature later; and among them were

1) A prediction of strong $3p - 3s$ gain in the selenium fluorine-like sequence near 204 Å at high incident laser intensity, but no experimental observation;

2) Experimental observations of dependence of gain of the observed 206 and 209 Å lines on laser pulse length, and whether irradiation was from one or both sides o the film, neither of which is explained by theoretical modeling;

3) A large difference between calculated and observed $3-3$ spectra down the laser axis for non-high gain transitions in the neon-like and fluorine-like sequences.

Theoretically there are a number of rather basic problems which are very much a mystery, and in time we hope that better understanding may come. It is not obvious in which of the many areas the problems lie, however, candidate areas include atomic physics, laser-plasma interaction, turbulence, propagation, resonant absorption, and line broadening.

In closing this section, we note that there is substantial effort in the area of Z-pinch implementation of the $3p - 3s$ approach [38-41], both in the areas of theory and experiment.

Future work will answer some of the questions which are currently outstanding in the field, and we may expect to see electron collisionally-pumped lasers operating at a number of laboratories around the world in coming years. Extension of the approach to higher shells is of interest; in Ref. 41 the use of Ni-like ions and $4d - 4p$ transitions is considered. There is no reason not to begin thinking about $5f - 5d$ transitions in many-times ionized ions with a closed N-shell.

III. Recombination Lasers

In 1964, Gudzenko and Shelepin published a paper discussing recombination in a decaying hydrogen plasma as an approach to the creation of population inversions between

Rydberg levels [43]. The basic idea is that if a hydrogen plasma is ionized in some manner, and then cools, population inversions form due to the more rapid decay of the lower members of the Rydberg series to the ground state. This mechanism can be used to invert Rydberg levels of low Z hydrogenic ions as well, and in this case, the level separation of the Rydberg levels is sufficiently large that the gain produced may be on EUV or soft X-ray transitions. There is no restriction of the scheme to hydrogenic ions (although most of the available literature is concerned with hydrogenic ions), and in principle inversions could be arranged through recombination pumping in any singly or multiply stripped ion.

Much of the material covered in this section has recently been reviewed by Boiko et al. (1984) [92].

There are several early works on the formation of inversions in hydrogen and helium plasmas (Ref. 43 - 48), and while the transitions examined are not in the EUV or soft X-ray regimes, this work is the predecessor of the more recent investigations into the shorter wavelength regimes. The early analysis of recombination in hydrogenic ions of Bates and co-workers [49–51] have played a very important role in the development of the field. Although these researchers were interested in astrophysical plasmas rather than laser kinetics, their work and further results based on their models scope out the parameter regimes wherein population inversions may occur.

Early theoretical analyses of the inversion kinetics in low Z hydrogenic ions were reported by Bohn (1974) [52], Pert and Ramsden (1974) [53] and Jones and Ali (1975) [54,55]. Although the models and assumptions differed between these analyses, the resulting picture was quite optimistic in terms of creating a short wavelength laser. On the experimental side, Irons and Peacock (1974) [56] reported measurements from which gains below 10^{-4} cm^{-1} were inferred on the hydrogenic carbon 3-2, 4-2, and 4-3 transitions. Dewhurst et al. (1976) [57] inferred a gain length product of about 0.02 on the 182 Å C VI 3-2 transition from experiments on expanding thin carbon fibers, a result in good agreement with theoretical modeling of Pert (1976) [58]. The peak gain in this case was calculated to peak near 0.6 cm^{-1}.

The 182 Å 3-2 line of C VI has been the most studied recombination-pumped short wavelength laser transition. Theoretical results and criticisms appear in Ref. 59-61, and C VI is central to all discussions. Key et al. (1976,1979) carried out experimental work on the irradiation of thin carbon fibers [62,63], and deduced a small signal gain of 0.2 cm^{-1} on the 182 Å line. Pert (1979) [64] extended his design calculations on gain in fiber expansion.

In 1981, Jacoby et al. published their experimental work on carbon fiber expansion, in which a gain length product of 5.0 was claimed for their best single result, and five fibers gave gain-length products between 2.5 and 3.5 [2]. The claims were based on measurements taken by nearly-matched spectrographs on- and off-axis observing spectra in the vicinity of the 182 Å 3-2 line. A much more detailed account of this work was presented in Ref. 65, including both experimental and theoretical results and discussion.

The demonstration of 5 gain lengths at 182 Å is an impressive result, and even reproduction of 2.5-3.5 gain lengths is very interesting. There has been some criticism (unpublished) of this work on the grounds that the 5 gain length product result is a single data point, and that neither the Hull group or any other group has managed to reproduce it. The same criticism applies to a lesser degree to the claims of 2.5-3.5 gain lengths, although in this case there are four data points, as well as further work at Rutherford [66,67], which appears to support the results. In addition, Pert and Shorrock (1983) [68] have recently reported observation of 3 gain-lengths at 134 Å on the 3-2 line in N VII (which is the analog to the 182 Å C VI line), on a carbon fiber with nitrogen impurity.

There are quite a number of works not yet mentioned which are concerned with inversions in hydrogenic systems (Ref. 69 - 82). 3-2 inversions at higher Z are examined in Ref. 69, and in Ref. 70 inversions agains the ground state are studied. Luk'yanov [71] and Jones and Ali [72] give results on the Z-dependence of recombination-pumped gain. Sato [73] examines He II recombination. In Ref. 74, a method for channeling expanding plasma into an elongated geometry suitable for EUV laser experiments is discussed. Zherikhin et al. [75] claimed observation of gain at on the 4-3 transition in C VI (of 6 cm^{-1}) and of intensity anomalies on the 2-3 and 2-4 transitions in Li-like sodium, the latter claim being argued as due to impurities by Elton and Dixon (1978) [83]. Khorukhov (1982) [76] measured oxygen K-shell emission from an aluminum oxide target, and inferred 4-3 gain on O VII of 0.1 cm^{-1}. In Ref. 77, experimental results are assessed in terms of progress towards development of significant gain on 4-3 transitions in hydrogenic and nearly hydrogenic ions. Dixon et al. (1983) [78] present experimental results for Rydberg levels in a laser-produced carbon plasma, and interpret the results in terms of an inferred C VI 4-3 gain of 0.01-0.02 cm^{-1}. Elton [79] comments on parameter regimes of interest for hydrogenic 3-2 and 4-3 recombination-pumped inversions. Boiko et al. [80] presented a variety of results for gain between M-, N- and O-shell states in hydrogenic fluorine produced by laser irradiation of a slab CF_2 target and observation of K-shell spectra. In Ref. 81, tables are presented giving results of gain calculations on a number of transitions of hydrogenic ions at various Z and comment on pumping requirements. Matthews et al. (1984) [82] present experimental results of measurements of the 102 Å 3-2 line of O VIII and indicate that the enhanced emission is consistent with a gain of 0.5 cm^{-1} (and gain-length product of about 0.6).

There are a number of works concerned with other recombination pumping in other isoelectronic sequences (Ref. 84-92). Bhagavatula and Yaakobi [84] monitored K-shell lines from helium-like aluminum and inferred a small signal gain of 10 cm^{-1} on the triplet $1s4f - 1s3d$ manifold at 129.7 Å , in a target geometry utilizing conductive cooling through the placement of a magnesium plate near the expanding aluminum plasma. Boiko et al. [85] also used K-shell diagnostics to infer inversions; however results were presented for the $1s6h - 1s5g$ singlet transition of helium-like magnesium at 620 Å . The inferred gain was reported to be a stunning 750 cm^{-1}. Zhizhan et al. [86] monitored K-shell lines from helium-like Mg and found a ratio of $1s-4p$ emission to $1s-3p$ emission of 4.3, and deduced that a population inversion occurred (no gain estimate was made) on the 154.8 Å 4-3 transition.

Lithium-like systems were studied in Ref. 87-90. Kononv and Koshelev [87] present theoretical results for 4-3 transitions in Li-like Al at 150 Å and predict gains of 0.1-1.0 cm^{-1}. Later experimental work [88] inferred a gain of about 0.1 cm^{-1} on the $4f-3d$ transition from observation of 2-3 and 2-4 emission. Gaponov et al. [89] examines a possible transient 3-2 inversion in lithium-like neon. Bunkin et al. [90] discusses possible mechanisms for the Na IX results of Zherikhin et al. [75] discussed above.

Silfvast et al. [92] discuss possibilities for extending recombination schemes in the tin and cadmium sequences into the EUV regimes.

Most of the work described so far involves freely expanding plasmas, in which the temperature drop required to drive the recombination is provided by hydrodynamic expansion and possible thermal conduction to a nearby heat sink. In a series of works by Suckewer and co-workers [93-97] the possibility of recombination-pumping in a magnetically confined plasma is explored. In this case radiative cooling provides the cooling requisite to drive the recombination. The major theoretical discussion is presented by Suckewer and Fishman (1980) [94], and in Ref. 95-97, published experimental observations of the 182 Å C VI line indicates a gain-length product of 3.5. More recent work not yet published shows even more amplification, for example in Thomsen's account of the Boston APS talk [97] we find a preliminary claim of 6.5 gain-lengths reported. This claim, if it stands the test of time as well as critical scrutiny, is potentially very significant.

The use of a confined plasma and recombination-pumping to drive EUV and soft X-ray laser inversions appears to be extremely promising, especially in view of the remarkable experimental progress made to date. To be able to maintain or even increase plasma density while at the same time drastically reducing electron temperature through radiative cooling appears to be a very powerful combination, and we anticipate further exciting developments in this area in the coming years. We note that even though the most promising experimental results presented so far are on the C VI 182 Å line, the technique is very general. In the above-cited works some discussion is devoted to inversions in low-Z lithium-like ions as well as C VI.

Before leaving the general area of recombination lasers, we note that a substantial body of work has been published by a group of French workers [98–106] on the subject of collisional and recombination schemes. Much of the work has concerned an intensity anomaly on a 117.4 Å line in an aluminum plasma, which is attributed to the $2p^5 4d\,^3P_1 - 2p^6\,^1S_0$ transition in neon-like aluminum. In Ref. 106, a discussion is given concerning recombination-pumping in lithium-like aluminum at 105.7 Å ($5f - 3d$ transition). The presence of gain at both 105.7 Å and 103.8 Å is deduced from a comparison of axial spectra taken at two sets of different lengths (4,5 mm and 7,9 mm), and a gain (in the absence on continuum absorption) of 0.5 - 2.0 cm^{-1} is estimated (the value in the presence of continuum absorption is much less).

IV. Photoionization Pumping

The first serious paper concerning the design of EUV and soft X-ray laser design which we have found was published by Duguay and Rentzepis in 1967 [107], and described possible short wavelength lasers pumped by photoionization. The basic idea is to remove inner-shell electrons selectively from either neutral atoms or ions which have lost a small number of electrons through one mechanism or another; upon such removal, the resulting population distribution may be inverted against radiative transitions filling the inner shell hole. The possibility of selective removal of inner-shell electrons (instead of optical electrons) occurs because inner-shell photoionization cross sections are usually much larger than those of outer-shell electrons, though the thresholds occur at substantially higher energy.

Duguay and Rentzepis suggested that sodium would make an excellent candidate in which to implement the scheme, as a $2p$ electron could be readily and selectively removed via X-ray pumping above the $2p$ edge at 38 eV. Unfortunately, once the inversion has developed, there is no mechanism to sustain it, and the lower laser state ultimately is irreversibly filled by the radiative decay of upper laser state population, or from other channels. The laser transition is self-terminating, and as such the requirements on the time allowed to pump an inversion is usually limited by the relaxation time between upper and lower laser states. For the sodium scheme, the radiative decay time was estimated to be 400 psec.

A second example was given, and that was the inversion of the $2p - 1s$ transition in singly-ionized Cu at 1.537 Å . Once again, selective inner-shell photoionization creates the inversion; however in this case the decay time of the upper state is about a femtosecond, the principal decay mode being Auger decay to doubly ionized species. After some longer time the lower laser state becomes populated, irreversibly, and the inversion is terminated.

Much of the work in the area is based on or follows rather directly from this early work. Some of the approaches in this and the following section are reviewed in Ref. 108.

Analyses of the K-shell approach were reported by Stankevich [109], Arecchi et al. [110], Elton [111,112], Axelrod [113], and Denne [114]. Duguay [115] presents results for the copper laser assuming delta function pumping. A modified version of the approach involving removal of several K-shell electrons was proposed in Ref. 116, discussed in Ref. 117, and analysed by Axelrod [118].

The L-shell approach and analogs were considered in Ref. 115, Ref. 119-125, and Ref. 12. Rozanov [119] pointed out that the approach was suitable alkali and alkali-earth elements in which case removal of a single M-shell or N-shell electron from a full shell would lead to analogous inversions if one or two optical electrons were present in the outermost shell. Jones and Ali pointed out the usefulness of radiation from an exploding wire in pumping the neutral sodium L-shell [120]. McGuire [121] pointed out that K-shell ionization in Na II followed by subsequent Auger decay would lead to $2p-2s$ inversions in the oxygen-like ion. The first experimental attempts reported for the photoionization approach were described by McGuire and Duguay [122], who for a number of reasons were unable to observe the effect.

Hagelstein [108] described an analog of the classical sodium scheme in which preferential $2s$ ionization from neutral neon would lead to a $2p-2s$ inversion in Ne II near 461 Å. Extensive analyses appear in Ref. 123 and 124, and further calculations are presented in Ref. 12. Some experimental work was carried out at the Omega facility at U. of Rochester (LLE) [125], including flashlamp measurements and attempts at measuring gain. No gain was observed on the Ne II $2p-2s$ transition on the 4 integral laser shots carried out.

Before closing our discussion of photoionization-pumped lasers, it is of interest to note some work of Csonka (Ref. 126-130). The proposal concerns the use of synchrotron radiation to pump through photoionization an inversion on the $1s^2-1s2p$ singlet transition of Li II at 199.3 Å. The idea is that lithium vapor (initially in a $1s^22s$ configuration) is optically pumped into the $1s^22p$ state, and then photoionized by K-shell ionizing radiation produced by a synchrotron. Transitions in other ions could be inverted transiently in a similar fashion [129].

Although these papers are concerned with direct transient pumping by the synchrotron radiation, which places a stringent requirement on pump brightness, it may be possible to reverse the order of the optical and synchrotron pumping. In such a scheme, ground state lithium ions could be pumped into the metastable $1s2s\,^1S_0$ state of Li II [126] over a long period of time, and then rapidly pumped into the $1s2p^1P_1$ upper laser state using an optical laser. In this case the scheme would be a metastable storage scheme, and we shall discuss the general class of such schemes in section VI.

V. Photoexcitation Lasers

The use of optical pumping with resonant line radiation to produce population inversions has been well-known to laser physicists for many years [131]. The first published proposal which we have concerning the extension of the approach into the EUV and soft X-ray regimes is that of Vinogradov, Sobel'man and Yukov (1975) [132], who proposed a number of candidate resonance matches between one- and two-electron ions of moderately low Z. The basic idea is that line radiation produced by a strong line (for example, a $1s-2p$ transition in a laser plasma) in the X-ray regime might be used to actively pump an X-ray transition in a different ion, such that upon strong pumping the level populations in the pumped ion are driven into an inversion. For K-shell / K-shell pairs at low Z, there is really only one match that is at all precise, and that is the Na X $1s2p^1P - 1s^2\,^1S$ transition at 11.003 Å as a pumping line, and the Ne IX $1s4p^1P - 1s^2\,^1S$ transition at 11.000 Å as the lasant line. Strong pumping of the $1-4$ transition of helium-like neon can in principle lead to an inversion of the $4-3$ singlet lines near 230 Å , and perhaps on the $4-2$ and $3-2$ singlet transitions near 58 Å and 82 Å as well.

Another proposed match between the helium-like ions of Si and Al has received much attention in the literature as well, although in this case the mismatch is several linewidths, such that additional measures must be taken in order to ensure that resonant pumping actually occurs. The particular pair of transitions involved are the Si XIII $1s2p^1P - 1s^2\,^1S$ transition at 6.650 Å as pump line, and the Al XII $1s3p^1P - 1s^2\,^1S$ transition at 6.635 Å as lasant line. Both of these low Z pairs were suggested in [132], as well as a number of others.

The principal difficulty with the photoexcitation approach in the EUV and soft X-ray regimes is finding precise resonances. Even though the approach is nearly a decade old, the number of confirmed precision resonance pairs which could be used profitably is dismally small, certainly less than 5. Much of the literature in the area is concerned with the issue of selecting and confirming resonances. The second major difficulty, given a precise match, is to arrange for the pump line to become usefully bright. This issue has also been a major area of interest in the literature.

In Ref. 133, it is proposed to use the C VI $2p-1s$ resonance line to pump the $1-4$ singlet line in C V. Due to the magnitude of the mismatch, it is suggested that additional broadening of the pump line could be arranged for through the use of a very optically thick pump line. Another proposal to systematically find resonances is to use hydrogenic ions, in which case a match is guaranteed (nonrelativistically) between $1-2$ pump lines of an ion with nuclear charge Z, and a $2-4$ transition of a lasant ion with nuclear charge 2Z. This systematic approach to the determination of resonances is appearantly due to Elton (unpublished, but later summarized in Ref. 134), and the first publication of the idea in the literature which we have found is by Bhagavatula (1976) [135]. The presence of an inversion in hydrogenic and helium-like magnesium ions was inferred experimentally [136] from observations of K-shell emission lines, and gave evdence that resonant photoexcitation from carbon (an implementation of the above mentioned Z, 2Z scheme) was indeed involved in the development of the inversion. This work and additional results are reviewed by Bhagavatula (1980) [137], and an inferred gain of 5 - 10 cm^{-1} was reported for the $4-3$ transition of hydrogenic Mg at 130 Å .

The Al / Si resonance scheme was studied by Apruzese et al. [138], who calculated $3-2$ gains in Al XII at 44 Å in the vicinity of 10^3 cm^{-1} when pumped by a hot, optically thick silicon plasma. This work was greatly extended in the detailed theoretical treatment of Whitney et al. (1980) [139], wherein a thorough examination of density and radiation parameters of interest in designing such a system were reviewed. Gains were found to be sensitive to pump radiation strength, and for the examples cited, reached values in excess of 50 cm^{-1}. Further results are given in Ref. 140. In the latter work is given a discussion of the dependence of gain on velocity in the case where the silicon and aluminum plasmas stream towards each other.

Other resonances were proposed and examined by Chapline and coworkers [141,142]. In Ref. 141, the problem of K-shell / K-shell resonances at higher Z is considered, and in Ref. 142 it is proposed to use K-shell and L-shell lines to pump $2-4$ transitions in mid-Z neon-like ions.

In Ref. 123, Hagelstein proposed using $3-2$ lines from L-shell ions (Li-like - Ne-like) to pump $1-3$ and $1-4$ transitions in one- and two-electron low Z ions. A list of over a dozen potential resonant pairs were suggested, but neither from theory or existing published experimental results could it be confirmed that any of the newly proposed candidate transitions were actually sufficiently precise to be of use. High resolution coincidence measurements were carried out at KMSF in Ann Arbor [143], giving results on a number of L-shell transitions. Of interest here are their results on a line in Be-like Mn near 12.641 Å [143] which is in proximity to the 12.643 Å $1s-3p$ lines of hydrogenic fluorine. Resonances were also reported in Cr and Mn near 14.458 Å , which are of interest in pumping the $1s^2\,{}^1S - 1s3p\,{}^1P$ transition of helium-like fluorine.

More recent work at KMSF which will be published shortly [144] was carried out at even higher resolution, and it was found that the Cr resonance near 14.458 Å is not quite as precise as reported earlier, but that the Be-like Mn line appears to match the hydrogenic fluorine doublet near 12.643 Å to within roughly a linewidth, or about one part in 10^4. Transmission experiments were attempted to measure the extinction of the Mn line in a fluorine-loaded plasma; experiments aimed at monitoring enhanced $2-1$ and $3-1$ emission from resonantly-pumped F IX were also performed.

As mentioned above, the brightness of a pump line is very important in terms of laser design. In Ref. 123, design simulations were based on the availability of pump lines with brightness 0.015 photons/mode, where the modal photon density $n(E)$ of a blackbody distribution at temperature T is given by [145]

$$n(E) = \frac{1}{exp(E/kT) - 1}$$

More recent design calculations [12,146,147] assume lower pump strengths in the neighborhood of 0.005 photons/mode. Experimental measurements of the strength of candidate pump lines from thin-film flashlamps are reported in Ref. 148 and 149, and the brightness of the B-like Cr line near 14.453 Å was estimated to be about 0.01 photons/mode. The pump brightness of the Mn line near 12.643 Å was examined in Ref. 13, and found to be 0.005 photons/mode (the recent measurements at KMSF of the line also contains information relating to brightness, and we may expect further reports on this in coming months).

The brightness of the helium-like sodium line was also measured [149], and found to be substantially weaker. This result is important in the design of lasers based on the Na X / Ne IX resonance discussed above (see Ref. 123, 140, and 12 for design calculations).

There have been two campaigns at LLNL aimed at directly measuring amplification from resonantly-pumped lasers [12,13,150], and as yet there is no convincing demonstration of amplification. Out of nine shots on the Mn / F resonance attempted on the second (1984) campaign, only two showed signs of early emission at 81 Å (the $3d^2D - 2p^2P$ F IX wavelength); although a potentially interesting result, it is clear that more hard work is required before amplification can be proven

Before, moving on to our next area, we note some interesting research of Trebes and Krishnan [151–153], wherein are reported observation of effects of photoexcitation in lower energy transitions. The use of Be-like ions as lasant ions is proposed in [153].

VI. Storage Lasers

The earliest proposal which we have found that fits into the general area of what we have termed "storage" lasers was published by Freund in 1974 [154], and concerned the development of a $2s - 1s$ inversion in hydrogenic oxygen and subsequent optical pumping of the $2s - 2p$ transition to obtain $2p - 1s$ gain at 18.97 Å . No mechanism was presented to develop the $2s - 1s$ inversion initially; rather the paper focussed on the details of the development of one- and two-photon gain in the presence of an optical laser field.

The basic idea behind storage lasers is to find a relatively long-lived (metastable) level which is highly excited, and which can be populated preferentially such that an inversion can be generated against lower state or ground state laser levels. Once the metastable state is inverted relative to the target lower laser state, then an optical laser pulse (or other means) may be used to rapidly dump population into a nearby upper laser state (and hence develop large gain), or else drive parametric gain via two (or more) photon processes. The major advantage of the approach is that the initial development of the inversion between the metastable level and the lower laser state may be carried out over a long time (and hence require low pumping power), and the stimulating optical laser pulse, which is usually of much shorter duration, may also be of low power.

Mahr and Roeder (1974) [155] were the first to focus on Li II as a candidate for a metastable scheme. In this case methods were suggested to obtain the initial inversion of the metastable $1s2s^1S$ level with respect to the ground state $1s^2\,^1S$, including photoionization and charge transfer. Other low Z ions were discussed briefly; however, then and now, lithium has played a central role in the area. Other early proposals by Gudzenko and co-workers [156,157] concerned storage in the helium-like triplet $1s2s$ state, and optical pumping into the $1s2p$ singlet or triplet levels.

A paper by Vekhov et al. (1975) [158] contains the earliest experimental results concerning metastable Li II level populations of interest in short wavelength laser studies. K-shell emission was monitored in a decaying lithium plasma produced by a high current discharge in lithium vapor, and the authors concluded that a considerable proportion of the ions were in the metastable level. No optical pumping was attemped.

Two papers by a group at AERL [159,160] considered an implementation of the Li II scheme, wherein X-rays from a laser-produced plasma would photoionize lithium vapor and produce a substantial population of metastable singlet and triplet $1s2s$ Li II ions. An optical laser pulse would the drive both one- and two-photon gain as discussed above.

We now turn our attention to a massive body of work by Harris and co-workers [161–175], who currently dominate this area. Metastable states and EUV transition work is almost synonymous with the research efforts of this Stanford group. An excellent review of this work is found in the present volume [161]; hence we shall only briefly summarize some of the key ideas. Some early work [162,163] is concerned with the creation of a very bright VUV source via Stokes and anti-Stokes sideband generation when an optical (Nd:YALG at 1.064 μ) laser was focused into a CW glow discharge. Although no inversion was attempted, the approach is relevant to our present discussions.

The key proposal around which much of the Stanford work is centered is a class of metastable storage schemes, the first of which was presented in Ref. 164. The basic idea is that electron collisional excitation in a moderately warm low density lithium plasma would result in substantial inner-shell excitation, and hence population of the $1s2sp\,^4P$ metastable states. Optical promotion of the metastable population to one of the $1s2p^2\,^2P$ upper laser states would then be accomplished using an optical laser pulse at 2949 Å . The EUV laser transitions would then be through a $2p - 1s$ transition down to the $1s^22p$ doublet lower laser states. In order to ensure the presence of an inversion, Harris proposes that the 2949 Å optical pump radiation would also photoionize the lower laser state, which otherwise might be populated thermally via electron excitation from the Li I ground state.

Due to the purity of coupling between doubly excited levels of Li I, the quartet to doublet optical transition has a very small oscillator strength, and hence imposes a very high power requirement on the optical pump laser. Additional proposals of Ref. 166,173 and 174 are concerned with schemes (both in lithium and in other neutral and singly ionized alkali systems) with a larger degree of configuration mixing such that the optical pump power requirements are reduced, while retaining metastability.

Caro et al. (1983,1984) [173,174] presents some interesting results from experimental work on the production of metastables from photoionization by laser-plasma X-rays. Detailed kinetics measurements were made on a number of states including the metastable Li II $1s2s$ states discussed above.

Even though most of the recent work in the area of storage lasers has been done by Harris and co-workers, we close this section with a few citations from elsewhere. In Ref. 176 is proposed and demonstrated experimentally a novel technique for producing Li $1s2sp\,^4P$ metastable neutrals by surface capture of electrons onto lithium ions during grazing incidence scattering from a ferromagnetic crystal. In Ref. 177 is a discussion of an alternative approach to the creation of alkali quartet metastables via collisional excitation transfer from rare gas metastable states.

VII. Charge Transfer Lasers

The earliest proposals for EUV and soft X-ray lasers which are pumped through the transfer of an electron from a donor atom or ion to an upper laser state are due (as far as we know) to Vinogradov and Sobel'man (1973) [178,179]. The basic idea was to irradiate a solid laser target which is surrounded by a buffer gas. Upon laser irradiation, the outer skin of the solid target would ablate off as a multiply ionized plasma, which in the presence of the buffer atoms could develop a population inversion through selective population of an upper laser state via charge transfer. Electrons from the cold buffer gas would be picked up by the ions of the expanding plasma, and the preferential population of upper laser states would be ensured by the approximate energy conservation of the charge transfer process.

The use of charge exchange as a mechanism to drive population inversions in the visible and UV had been studied earlier [180–183]. The use of the charge exchange mechanism to remove an inner-shell electron (as opposed to donating an electron to an outer shell) from the lasing ion was discussed early on by McCorkle [184,185], but has not been discussed much in the more recent literature. The main efforts reported so far have been concerned with the more classical approach of charge donation into an upper laser state.

Substantial theoretical work on charge transfer EUV lasers was reported in a series of works by Scully and co-workers [186–189]. The scheme was to be implemented by passing an ion beam of bare helium (or other species) ions through a thin-film solid or gas jet hydrogen (or other donator) target. The principal example discussed was the inversion of the $2p-1s$ transition at 304 Å in He II after electron pickup from hydrogen. An extensive review of this work is given by Cantrell et al. [190].

There is a proposal and analysis of the use of a shock tube to generate lasant ions and to provide an interaction region wherein mixing might occur [191].

Elton [192] reviewed the prospects for a laser-plasma implementation of the approach, which corresponds more closely to the early proposals of Vinogradov and Sobel'man [178,179]. Dixon and Elton [193] reported the first experimental results implicating charge transfer as a mechanism for enhancing N-shell population in lithium-like and helium-like carbon. Interestingly enough, their results showed effects in the presence of a background gas (as opposed to vacuum), however, there was no particular dependence of the magnitude of the effect on which background gas was employed or what pressure was used. Some conjecture was given as to the source of the transferred charges, including lower ionization stages of carbon within the blowoff plasma. Theoretical work and further discussion is given in Ref. 194. The presence of fast neutral carbon atoms preceding and during the ablation of ionized carbon plasma was demonstrated experimentally in Ref. 195.

There are a number of papers by a group at Cornell [196–200], who observed charge exchange in a variety of systems resulting in strong VUV fluorescence. The initial proposals [196,197] concerned charge transfer from neutral cesium to protons, resulting in a transient $2p-1s$ inversion at 1215.67 Å . Observation of charge transfer from neutral cesium and excited neutral sodium to protons was reported in Ref. 198 (transfer from cesium had been previously studied; see Ref. 201 and citations therein), and for other systems in Ref. 199. A direct measurement of gain on the $2p-1s$ transition of H I was reported by Tkach et al. (1980) [200], with a gain coefficient of 1.4 cm^{-1} being reported.

There is a paper by Fukuda and Suemitsu [202] of observation of $2p-2s$ transitions in low Z impurity ions in a linear Z-pinch helium plasma, from which it was concluded that charge transfer of $2s$ electrons to He II ions played an important role in line formation. The authors proposed to use the removal of $2s$ electrons as a mechanism to invert $2p-2s$ transitions and construct a VUV laser.

VIII. Free Electron Lasers in the EUV and Soft X-ray Regime

A number of papers have appeared quite recently concerning proposals for free electron lasers in the VUV, EUV, soft X-ray and X-ray regimes [203–206]. As the area is so new, and as our references are so incomplete, we are not in a position to speculate on which proposal was first. It seems that the time is ripe for work in the area, and we expect the field to blossom in coming years.

In Ref. 203 and 204, the operation of free electron lasers in the VUV, EUV and soft X-ray regimes is discussed. The proposals concern the use of magnetic undulators with a few centimeter spatial frequency to modulate a high energy (GeV) electron storage ring [203] or a high energy (200 MeV) electron beam from a linac [204]. The use of a storage ring and magnetic undulator is also discussed in Ref. 205.

The extension of the approach to higher laser energy is inhibited by constraints placed on the magnetic undulators, and in Ref. 206 and 207 appear proposals for an optical wiggler [206] to modulate a 5 MeV beam to obtain gain near 5 Å , and for modulation of the dielectric constant in striated media [207] to produce gain in the X-ray regime (for example, at 3 Å from a 60 MeV beam).

IX. Other Methods

There are a number of proposals which do not fit well into the catagories which we have discussed in previous sections, either due to the proposal of a novel pumping mechanism or type of laser transition. In this section we shall attempt to survey some of this work.

One example of a novel laser transition is the proposal to use radiative autoionization as a mechanism with which to construct a new class of laser [208,209]. The idea is to create very highly excited metastable atoms or ions, for example the He^- $2p^3\,{}^4S^0$ quartet metastable state, whose principal decay mode involves the spontaneous emission of a photon and the ejection of an electron (radiative autoionization, RA). In the case of the quartet metastable helium negative ion, the RA decay is to the $1s2p\,{}^3P$ state of He I, accompanied by the ejection of a p free electron. The spontaneous radiation which is emitted is very sharply peaked around 323.15 Å in the EUV. There are evidently quite a few systems available which have suitable properties, both with respect to metastability and strength of the radiative autoionization effect, to be candidates for laser transitions, including the $2p^2\,{}^3P$ triplet metastable of He I and H^- and the $1s2p^2\,{}^2P$ states of Li I and Be II [209].

The molecular analog of radiative autoionization is the radiative decay of a bound state to a dissociating lower molecular state, and constitutes the physical mechanism for the operation of optical excimer lasers.

Another approach is based on on the observation that the asymmetry of absorption line profiles in the case of where the upper state is autoionizing means that gain may be obtained even in the absence of a population inversion (Ref. 210).

Miller [211] suggests the use of an accelerated atomic beam (for example, atomic hydrogen), and exploitation of the Doppler shift between forward and backward travelling waves. Optical light directed into the beam would be upshifted in the frame of the fast-moving hydrogen, and if with the moving frame the optical pulse is circularly polarized and short such that the lower $1s$ state population is coherently raised to the upper $2p$ state, then it is speculated that superradiance might be arranged within the atomic beam. The resulting forward wave would then be upshifted in the laboratory frame, and with sufficiently high accelerator energies, be shifted into the X-ray regime.

Electron tunneling was proposed as a mechanism to produce a K-shell vacancy in an early paper by Lax and Guenther (1972) [212]. This approach is most closely related to the early K-shell photoionization schemes discussed in section IV.

For historical reference, there was a report by Kepros et al. (1972) of observation of lasing in a $CuSO_4$ doped gelatin which was irradiated with a Nd laser [213]. This work received much criticism (see the discussion in the review paper of Elton et al. [8]) and is currently held in low regard by many XRL researchers

There is a large body of work in the literature concerning the development of gamma-ray lasers, which is out of the scope of the present review. Interested readers are referred to the excellent recent reviews of Baldwin, Solem and Gol'danskii (1981) [214] and Baldwin (1982) [215].

IX. Mirrors and Cavities

No review of short wavelength lasers can be complete without at least some discussion of EUV and soft X-ray reflective optics. In spite of the fact that much of the literature on approaches is concerned with amplified spontaneous noise systems, lossy mirrors have been demonstrated in the EUV and soft x-ray regimes, and future experimental work is likely to rely much more heavily on mirrors, cavities and optics than that reported to date.

There are a number of early proposals for X-ray laser cavities [216–219], which are based on the extreme efficiency of Bragg reflection in the X-ray regime. Experimental peak reflectivities in excess of 0.95 had been reported for a copper K-shell line at 1.537 Å on near-perfect planar germanium crystals [220], and hence the prospects for very high quality X-ray laser resonators exploiting Bragg reflection are promising.

Unfortunately, as discussed in previous sections of this review, most current serious short wavelength laser work is focused on EUV and soft X-ray regimes, for which natural crystal Bragg reflection is not particularly attractive. Current proposals are based on artificial multi-layered mirrors, which we shall discuss shortly. A recent review of EUV reflective optics is given by Spiller (1984) [221].

The cavity used by Ilyukhin et al. (1977) [1] relied on high reflectivity from four simple metallic surface reflectors. Normal incidence reflectors work reasonably well down to about 300 Å [222]; below this wavelength there are no satisfactory normal incidence non-layered mirrors available. Glancing angle or surface wave cavities [223,224] are still a possibility, although there are often constraints placed on this type of approach due to the finite and oftentimes short duration of the EUV or soft X-ray population inversion.

There has been substantial work done on multilayer soap crystals [225–227], which have interesting reflective properties in the EUV and soft X-ray regimes. The idea is that long chain fatty acids can form lipid bilayers, and with work, many bilayers can be arranged on top of one another to form a multilayer artificial crystal. If the fatty acid has a high Z atom (for example, lead) attached at the carboxyl end, then in the multilayer crystal, the heavy atoms will be aligned in planes in the middle of each bilayer.

More efficient reflecting structures are made from artificial multilayers of various compositions, usually alternating layers of high and low density films, so as to maximize the spatial modulation of both real and imaginary dielectric constants [228–240]. The maximum reflectivity obtainable is strongly dependent on the wavelength required and mirror components. Vinogradov and Zel'dovich (1976) [233] give theoretical results for a number of possible designs, with peak reflectivities in the neighborhood of 0.10 - 0.30 over much of the EUV and soxt X-ray regime. For example, at 182 Å (which is of interest in a number of C VI schemes), a reflectivity of 0.28 is calculated for an aluminum/bismuth multilayer mirror. At 81 Å (F IX $3-2$), a reflectivity of about 0.40 is theoretically attainable using a carbon/gold multilayer reflector. Experimental demonstration of substantial reflectivity at near-normal incidence is reported in Ref. 236-240.

The multilayer approach to high reflectance optics seems to be most promising in the EUV and soft X-ray regimes. The collisionally-pumped Se experiments carried out recently at LLNL [4] included some shots where multilayer mirrors were employed to increase output. Although the results were not dramatic, there was some increase in output which may be attributable to reflection.

Before closing this section, we note some early proposals for distributed feedback cavities for X-ray lasers [241–245]. Although most of the proposals were for crystalline cavities, and hence of use for X-rays, Yariv and Yeh [245] are concerned with an implementation in artificially layered media at longer wavelength. There remains the problem of density, as the proposals are for solid distributed feedback cavities while most of the major approaches to short wavelength lasers currently involve lower density plasmas or gases.

XI. Summary and Conclusions

We have reviewed the major approaches which have been suggested for the development of EUV and soft X-ray lasers. The field is currently quite exciting and promises to become more so in coming years. Direct observation of several gain lengths in the EUV has been reported now by four groups, using electron collisional [1,4] and recombination [2,3] schemes. Prospects for the development of resonantly-pumped lasers appear promising, especially with the advent of a confirmed high-precision resonance between Be-like manganese and H-like F near 12.643 Å [144], where the prospective pump line is quite bright [13]. The significant recent advances in the understanding and technology [164–174] of the quartet metastable approach to EUV storage lasers hold promise for the actual development of lasing systems in coming years. The recent development of high quality EUV and soft X-ray reflective optics [236–240] is a very significant occurrance in the field and we may expect to see much more of their use in short wavelength laser experiments in coming years.

XII. Addendum

No matter what efforts one goes to in compiling references for a review, after it is written, one or two works end up being left out through inadvertence. In this case, there is an important paper of Dixon and Elton (1984) [246] on collisional and resonant excitation schemes which is of interest in sections II ad V. We also note a paper of De Martini and Edighoffer (1980) [247] on short wavelength lasers via optical klystron, which is relevant to section VIII. A nice early review of soft X-ray lasers is given in Ref. 248. Conturie et al. [249] gives both theoretical and experimental results on a recombination scheme in helium-like aluminum. Bellum et al (1982) [250] examines charge exchange between cesium and ionized helium.

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